Femtosecond Structural Dynamics

Time-Resolved X-Ray Diffraction of Laser-Excited InSb

A group from Berkeley Lab, the University of California, Berkeley, and the University of Aarhus in Denmark has measured ultrafast structural changes in indium antimonide by means of time-resolved x-ray diffraction with a novel femtosecond x-ray source at the Advanced Light Source (ALS). The experiments quantitatively resolved lattice expansion occurring over tens of picoseconds after excitation with a high-power infrared laser and established for the first time a 10-picoseond delay in the onset of the expansion. They also demonstrated the subpicosecond creation of a disordered region near the surface.

Femtosecond x-ray experiments are an emerging area of research in chemistry, solid-state physics, and biology. Dynamic processes in condensed matter occur on a time scale dictated by the period of a lattice vibration, typically about 100 femtoseconds. The adaptation of established techniques for determining atomic structure, such as x-ray diffraction and x-ray absorption fine structure could provide direct information about structural changes on this time scale, but the temporal resolution is limited by the pulse length of the x-ray source. For example, the ALS emits pulses lasting 30 picoseconds.

A number of ways to circumvent this limitation are under investigation at the ALS. The Beam Test Facility (BTF) makes use of the electron beam in the ALS linear accelerator rather than in the storage ring. In brief, firing a high-power infrared laser across the tightly focused electron beam at 90° generates femtosecond x-ray pulses by the process of Thomson scattering. In the present experiments with a 100-femtosecond titanium-sapphire laser, 50-MeV electrons in the linac resulted in 300-femtosecond pulses of x rays with a photon energy of 30 keV and 25-MeV electrons in 500-femtosecond x rays at 7.3 keV.

The Berkeley group used the BTF source to measure x-ray diffraction from the (111) lattice planes of an indium antimonide crystal at intervals of a few picoseconds both before and after laser irradiation at intensities below the threshold for damage. Diffraction data for 30-keV photons revealed broadening of the diffraction peak beginning 10 picoseconds after laser irradiation, accompanied by a shift of the peak to lower photon energies and an increase in diffraction intensity. Measurements with 7.3-keV photons were more surface sensitive and exhibited a rapid decrease in the diffracted intensity in less than 1 picosecond, followed by a peak shift after 10 picoseconds similar to that at the higher photon energy.

These data demonstrate that onset of expansion of the lattice, which shifts the diffraction peak to lower photon energies, occurs only after a delay of about 10 picoseconds after laser irradiation. This is the first direct evidence of such a delay, which the researchers quantitatively modeled as a combination of energy-relaxation processes by which energy is transferred from the electrons created by absorption of laser light to lattice vibrations and propagation of the resulting lattice expansion inward from the surface. The decrease in diffraction intensity from the surface is due to the high concentration of photoexcited electrons, which cause a non-thermal disordering of the lattice on a time scale too short to invoke transfer of energy to lattice vibrations and subsequent bond breaking.

In collaboration with researchers from the Berkeley Lab Center for Beam Physics and the ALS, the Berkeley group is building a new bend-magnet beamline (Beamline 5.3.1) that will provide 100-femtosecond x-rays pulses using another laser-based technique, recently demonstrated at the ALS, called time slicing. In addition to the improved time resolution, this beamline will be about 10,000 times brighter than the BTF source and will cover the entire spectral range of the bend magnet, thereby opening a wide range of scientific applications for time-resolved x-ray measurements.

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A. H. Chin, R. W. Schoenlein, T. E. Glover, P. Balling, W. P. Leemans, and C. V. Shank, "Ultrafast structural dynamics in InSb probed by time-resolved x-ray diffraction," *Phys. Rev. Lett.* **83** (1999) 336.

RESEARCH FUNDING: Division of Materials Sciences (DMS), U. S. Department of Energy; National Science Foundation. Operation of the ALS is supported by DMS.

FEMTOSECOND STRUCTURAL DYNAMICS

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- **X-ray studies of ultrafast processes in condensed matter**
  - *Emerging area of research in physics, chemistry, and biology*
  - *Fundamental atomic motion on time scale of a lattice vibration (100 fs)*
- **Beam Test Facility at the Advanced Light Source**
  - *Electron beam from ALS linac interacts with femtosecond infrared laser*
  - *Thomson scattering generates 300-femtosecond x-ray pulses*
- **Berkeley group experiments with indium antimonide**
  - *Lattice expands over tens of picoseconds after laser irradiation*
  - *First direct observation of delay in onset of expansion*
  - *Disordered region formed at surface in less than 1 picosecond*
- **New bend-magnet beamline will extend capabilities**
  - *Laser-based time-slicing technique demonstrated at the ALS*
  - *X-ray pulses across the full bend-magnet spectrum*
  - *100 fs time resolution with10,000 times higher x-ray brightness*

R.W. Schoenlein, T.E. Glover, and W.P. Leemans (Berkeley Lab); A.H Chin and C.V. Shank (University of California, Berkeley, and Berkeley Lab); and P. Balling (University of Aarhus, Denmark). [*Phys. Rev. Lett.* 83 (1999) 336]

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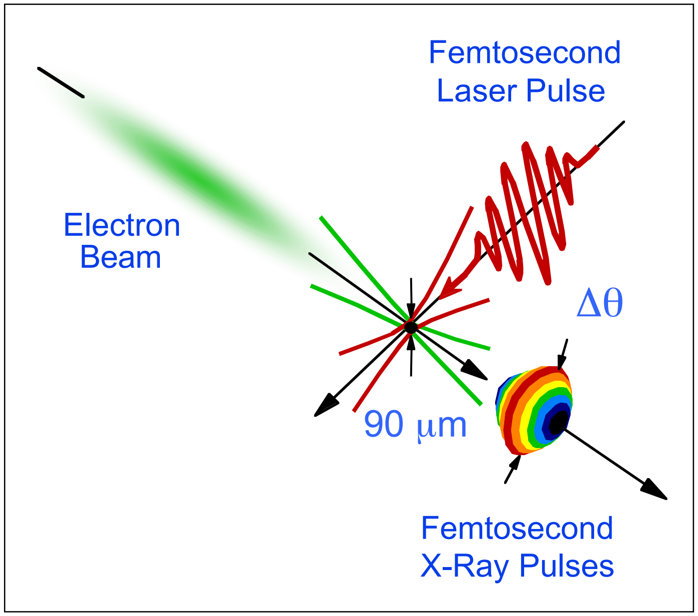

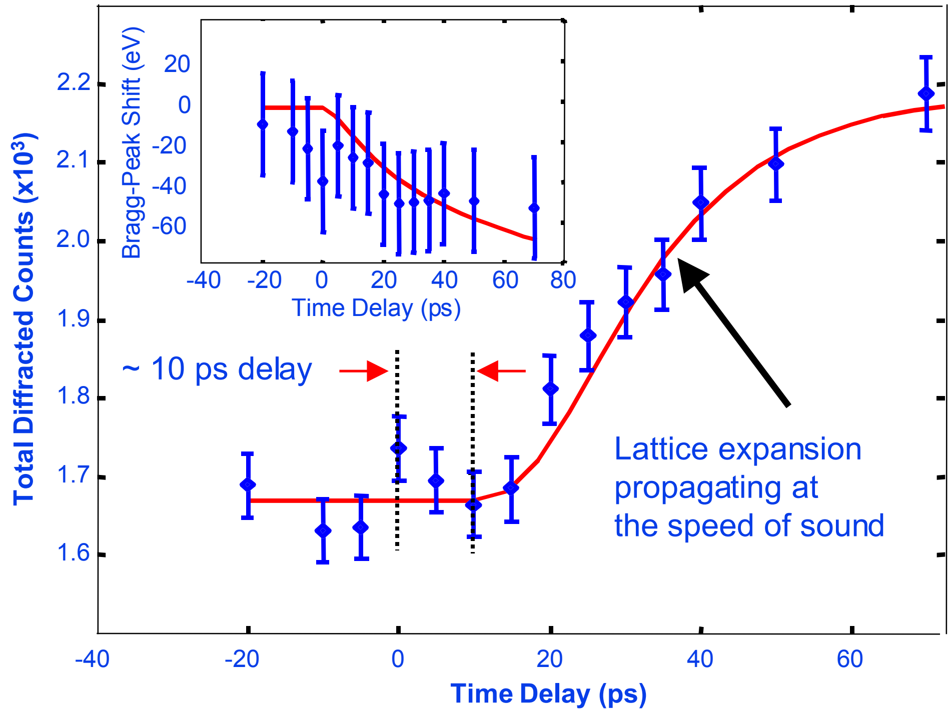

Firing a laser at 90° across a focused electron beam generates femtosecond pulses of x rays by Thomson scattering.

Shifts in Bragg peak and intensity of diffracted x rays show that laser heating of InSb results in rapid lattice expansion after a 10-ps delay.

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